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Author(s)	安徳, 重敏; 吉永, 春馬
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THE MEASUREMENT OF T(d,n) REACTION NEUTRON FLUX BY

ACTIVATION METHOD AND SCINTILLATION METHOD

By

Shigetoshi Antoku and Haruma Yoshinaga

Department of Radiation Biology, Research Institute for

Nuclear Medicine and Biology, Hiroshima University

Hiroshima, Japan

(Director: Prof. Dr. H. Yoshinaga)

放射化法およびシンチレーション法による T (d,n)反応中性子線束の測定

広島大学原爆放射能医学研究所障害基礎研究部門

安　徳　重　敏　吉　永　春　馬

S(n,p) P反応およびプラスチックシンチレータを用いて14MeV 中性子の線束測定を行つた.

S(n,p) P反応による相対的線束測定の精度は±10%以内に納まるが，絶対測定を行う場合は14 MeV 中性子に対するS(n,p) P核反応断面積として 0.3と0.37バーンの2つの値が知られており，いづれの値を用いるかによつて得られる数値が異なる.

水素を含有するプラスチック・シンチレータを用いたシンチレーション法は水素の核反応断面積が充分に信頼出来る値であるので反跳炭素核およびγ線による影響を除出すれば，かなりの精度で中性子線束を測定出来る．本研究で用いたシンチレータ（1″×1″φ）の計数効率は 14MeV 中性子に対して，1回衝突のみとして計算すれば0.0795/cm²，2回衝突を考慮に入れると0.0859/cm²であつた．この様に計数効率を知つておけば通常の測定に手軽に且つ簡単に使用出来る特長を持つている.

S (n,p) 反応で得られた数値に対するシンチレーション法の数値の値の比は0.86〜1.14の範囲にあり，ほゞ1に近い値が得られた.

Introduction

For the protection against radiation arising with the increasing utilization of nuclear equipment and for the determination of the maximum permissible dose, it is important to study the biological effects and the relative biological effectiveness of neutrons on animals. The reliability of the results obtained in these experiment mainly depends on the biological conditions and on the determination of neutron dose. Particularly the effect of the latter is greater.

To measure the neutron flux, ionization chamber, long counter, plastic scintillator and activation method have in general been widely used.

In our laboratory, the activation method using $^{32}S(n,p)^{32}P$ reaction and the plastic scintillation method are being used to measure neutron flux. The present paper described the calculation of neutron flux based on the counts recorded by the scintillation spectrometer equipped with plastic scintillator and on the amount of radioactive products, and also discusses the identification of radioactive products in neutron irradiated sulfur and the comparison of the data obtained by the two methods.

I Radioactivation method

The neutron source used in the study is T (d,n) reaction neutron generator. This generator supplies monoenergetic neutrons with 14.1 MeV. In order to measure the flux of 14.1 MeV neutrons by radioactivation technique, there are several useful nuclear reactions which produce radioactive products. However, nuclear reactions which give rise to radioactive products with short half-lives are not preferable, because the density of neutron flux generated by this apparatus is not always constant and particularly in long term irradiation, it decreases gradually with the decrease of the tritium target. When radioective products have long half-lives, the disintegration of radioctive products during neutron irradiation can be ignored and even if the competing reaction occurs, the by-product can be removed from the sample by chemical procedure.

As the nuclear reaction cross section of $^{32}S(n,p)^{32}P$ reaction for 14.1 MeV neutrons is rather large and the resultant radioactive phosphorous ^{32}P has a long half-life, this reaction is a useful detector for the measurement of neutron flux.

a) Materials and Methods

Fast neutrons were produced by bombarding a tritium target adsorbed in thin titanium layer with deuterons accelerated at 150 kV(maximum accelerating voltage: 200 kV, full load: 1 mA).[1]

Sulfur (reagent grade) was recrystalized twice from carbon disulfide containing activated charcoal and then powdered and packed in polyethylene vessel (1.2 cm in diameter and 3.5 cm in height). The irradiated sample was transferred to a stainless steel dish and burnt in a sand bath. The activity of the residue was measured by a Geiger-Müller counter and the amount of radioactive phosphorous ^{32}P produced by neutron irradiation was determined by comparison with the activity of standard ^{32}P. The integration of the neutron flux density of the entire exposure period can be obtained by the equation (1)

$$A = \frac{\tau \cdot W \cdot N}{A_s} \cdot f \cdot \sigma_s \left(1 - e^{-\frac{0.693}{T} t}\right) \quad \cdots\cdots\cdots\cdots(1)$$

where A is the disintegration rate of ^{32}P produced; A_s, the mass number of sulfur (=32); τ, the abundunce of ^{32}S in natural sulfur (=0.95); W, the weight of sample; N, Avogadro's number(=6.02×10^{23}); f, the neutron flux density in unit time; σ_s, the nuclear reaction cross section of $^{32}S(n,p)^{32}P$ reaction for 14.1 MeV neutrons (=0.30 or 0.37 barns); T, the half-life of ^{32}P(=14.3 days) and t, irradiation time.

As the two values of nuclear reaction cross section of $^{32}S(n,p)^{32}P$ reaction, 0.3[2] and 0.37[3] barns are give in literature. Tochiline et al[4] and Fujita et al[5] used the former

value, but there is no data to indicate which value is more accurate. Therefore, in this study the nutron flux was calculated by using both values.

b) Experimental results

Identification of activated products in neutron irradiated sulfur

The decay curves of the radioactive products are shown in Figs. 1 and 2. By analyzing these curves, the radioactive products were found to be a mixture of radioactive nuclide with a half-life of 140 to 180 minutes and that with a half-life of 14.2 to 14.4 days.

1) Radioactive product with the half-life of 14.2 to 14.4 days

In view of the half-life, the attenuation in Al plate and the energy distribution, this was found to be radioactive phosphorous ^{32}P produced by the $^{32}S(n,p)^{32}P$ reaction. As seen in Fig. 1, the half-life of the product is 14.2 to 14.4 days which agrees with that of ^{32}P (14.3 days in literature).
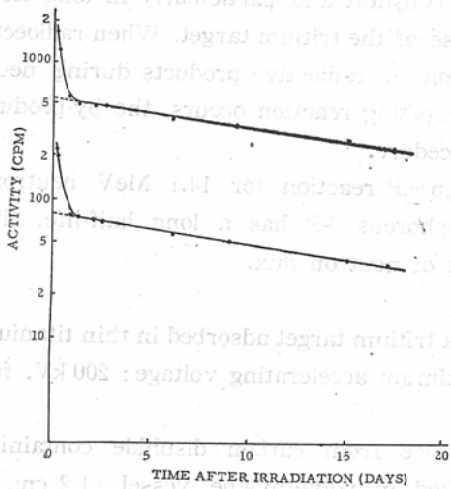

Figure 1 Two examples of decay curves of radioactive products in neutron irradiated sulfur
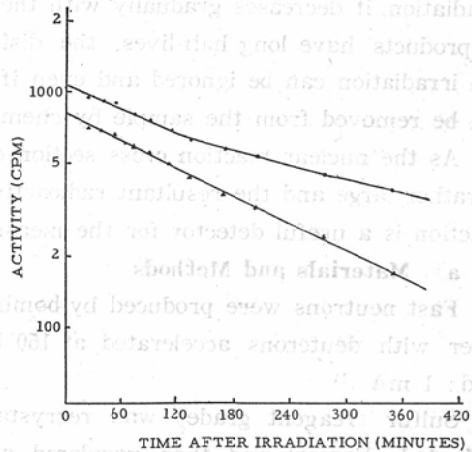
Figure 2 Decay curves of radioactive products in neutron irradiated sulfur
●: Total decay curve
▲: Decay curve obtained subtracting the activity one day after irradiation from the total decay curve
This is the decay curve of the radioactive product with short half-life

Fig. 3 shows the attenuation curve of the radioactive product in Al plates and that of ^{32}P. The activity of the product and ^{32}P attenuates with nearly the same behaviour. The half-value layer and maximum range of ^{32}P in Al plates have been reported to be 110 mg/cm² and 750 to 800 mg/cm², respectively. In this experiment, however, these were 96 mg/cm² and 700 mg/cm², respectively, which were somewhat low. The low energy of β-radiation seems to be attributed to the addition of back scattered radiation in the measurement of the activity on a stainless steel dish.[6] According to Feather's analysis, the maximum range of the product was 750 to 800 mg/cm².

Energy distribution curves of the product and ^{32}P determined on a stainless steel dish
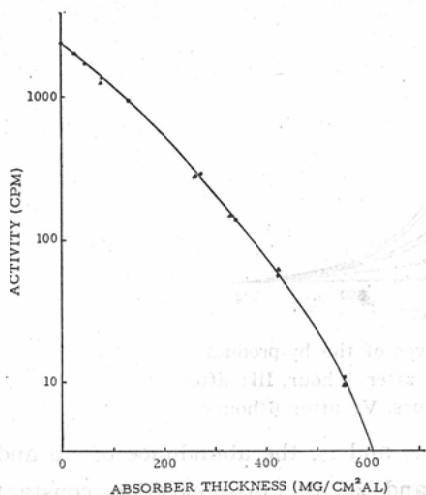
Figure 3 Attenuation curve of the radioactive
          product about one day after irradiat-
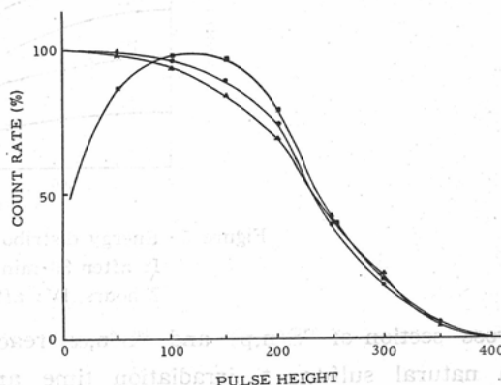   ion and ^{32}P in Al plates
          ●: Radioactive product
          ▲: ^{32}P

Figure 4 Energy distribution curves of the ra-
          dioactive Product about one day after
          irradiation, ^{32}P on stainless steel dish
          and ^{32}P in filter paper
          ▲: Radioactive product
          ●: ^{32}P on stainless steel dish
          ■: ^{32}P in filter paper

and that of ^{32}P adsorbed in a filtrer paper are shown in Fig. 4. The radioactive product
has an energy spectrum similar to ^{32}P. As seen in the attenuation curve, lower energy
radiation was found to be increased in radioactive substances on the stainless steel dish.

  2) Radioactive product with the half-life of 140 to 180 minutes

    Although Fujita et al[5] have been reported that there were no competing reactions
in ^{32}S(n,p)^{32}P reaction, in our experiment the competing reaction was observed and the
half-life of the by-product was 140 to 180 minutes. In Radiological Health Handbook[7]
and Neutron Cross Section[2], the description on ^{34}S(n,α)^{31}Si reaction is given. The nuclear
reaction cross section of the reaction for 14.1 MeV neutrons is 0.135 barns, the half-life
of ^{31}Si is 150 to 170 minutes and maximum energy of β-ray is 1.5 MeV.

    The half-life of the by-product almost coincides with that of ^{31}Si (see Fig. 2).

    Fig. 5 shows the energy spectra of the by-product which were obtained subtracting
from the energy spectra at given times after irradiation the energy spectrum 24 hours
after irradiation when the activity of the by-product is felt to be negligible. Although
it was felt that the by-product was a β-radiation emitter similar to ^{31}Si, identification
could not be confirmed by the energy distribution curve.

    The theoretical disintegration ratio of ^{32}P to ^{31}Si produced may be calculated by the
following equation

$$R = \frac{A_2 \cdot \tau_1 \cdot \sigma_1 (1-e^{-\lambda_1 t})}{A_1 \cdot \tau_2 \cdot \sigma_2 (1-e^{-\lambda_2 t})} \cdots\cdots\cdots\cdots (2)$$

where A_1 and A_2 are the mass number of ^{32}S and ^{34}S; σ_1 and σ_2, the nuclear reaction
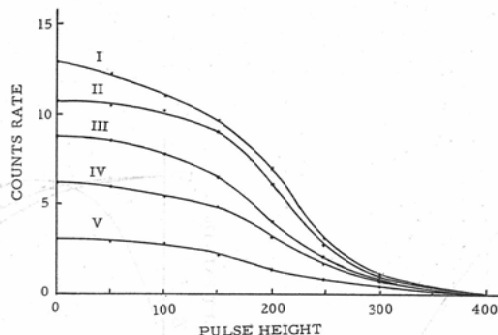
Figure 5　Energy distribution curves of the by-product
I: after 30 minutes, II: after 1 hour, III: after
2 hours, IV: after 3 hours, V: after 6 hours

cross section of $^{32}S(n,p)$ and $^{34}S(n,\alpha)$ reactions; τ_1 and τ_2, the abundunce of ^{32}S and ^{34}S in natural sulfur; t, irradiation time and λ_1 and λ_2, the disintegration constant of ^{32}P and ^{31}Si.

Shortly after irradiation, R was calculated to be one half from equatiσ^2on (2). In the calculation, the factors emplyed are: 1) 0.30 and 0.135 barns for σ_1 and σ_2; 2) 0.95 and 0.04 for τ_1 and τ_2; 3) 32 and 34 for A_1 and A_2; 4) 40 minutes for t; 5) 3.0×10^4 and 2.3×10^2 for λ_1 and λ_2.

In this experiment, the ratio of the main product, ^{32}P, to the by-product was about one fourth. The reason for the disagreement has not yet been determined. Further identification of the by-product will be made by chemical procedure.

Neutron flux measurement

In the preliminary experiment, it was determined that the counting efficiency of the G-M counter used was 8.24%. The determination of the counting efficiency was performed using standard radioactive phosphorous ^{32}P corrected by Kawada of the Japanese Electrotechnical Laboratory. The reliability of this source was $\pm 1.5\%$.

The activity loss from burning the sulfur and the effect of the sulfur amount on the activity loss was negligible. Fujita et al[5] have made a detailed report on the burning procedure and the extraction of NH_4OH.

The measurement of the activity was carried out 24 hours after irradiation and the amount of ^{32}P produced was determined by extrapolation to the period immediately after irradiation.

The measurement data of the neutron flux at 10 cm from the target are shown in Table I. The difference of each sample is less than $\pm 10\%$. Table II shows the total neurons generated at the target per second per 4π calculated from the activity of the samples at given distances from the target. The neutron flux density was calculated assuming that it attenuates by the inverse square of distannce from the target. As seen in Table II, the total neutrons generated at the target may be measured with considerable accuracy.

The scintillation doses in Tables I and II show the flux obtained by the scintillation

Table I Neutron flux calculated from activity of sulfur detectors at 10 cm from target(Irradiation time 60 min, σs = 0.3 barns)

Scintillation dose, 1.35×10^9 n/cm²		Scintillation dose, 1.56×10^9 n/cm²	
Sample No.	Neutron flux (n/cm²)	Sample No.	Neutron flux (n/cm²)
1	1.4×10^9	5	1.5×10^9
2	1.4×10^9	6	1.6×10^9
3	1.5×10^9	7	1.5×10^9
4	1.3×10^9	8	1.6×10^9
Mean ± σ	$(1.4 \pm 0.1) \times 10^9$	Mean ± σ	$(1.6 \pm 0.1) \times 10^9$

Table II Total neutrons generated per second per π, calculated from activity of samples at given distances from target, scintillation dose : single collision, activation : $\sigma_s = 0.30$ barns

Scintillation dose (n/sec/4π)	Target-sulfur distance (cm)	Neutron flux (n/sec/ 4π)
5.37×10^8	5.0	5.91×10^8
	8.2	5.87 〃
	10.2	5.54 〃
	M± σ	$(5.77 \pm 0.29) \times 10^8$
8.35×10^8	5.0	8.82×10^8
	8.2	8.64 〃
	11.0	9.46 〃
	M± σ	$(8.97 \pm 0.43) \times 10^8$
3.65×10^9	7.4	4.73×10^9
	〃	4.22 〃
	〃	4.03 〃
	10.6	4.56 〃
	〃	4.07 〃
	M± σ	$(4.32 \pm 0.31) \times 10^9$
2.42×10^9	7.4	2.38×10^9
	〃	2.40 〃
	〃	2.41 〃
	10.6	2.34 〃
	〃	2.49 〃
	M± σ	$(2.40 \pm 0.02) \times 10^9$
6.50×10^9	7.4	6.69×10^9
	〃	7.47 〃
	10.6	7.00 〃
	〃	6.95 〃
	〃	7.06 〃
	〃	6.69 〃
	M± σ	$(6.98 \pm 0.26) \times 10^9$

method. This technique will be described later.

II) Scintillation method[8]

In the last several years scintillation counters have been widely used for the detection of charged particles and γ-rays. Scintillators containing hydrogen can also be used to detect fast neutrons, since neutrons can produced recoil protons in the scintillator itself. The efficiency of such neutron detectors is conveniently defined as the ratio of the number of recoil protons to the number of incident neutrons. In practice, this efficiency cannot be used directly to convert the observed counting rate to neutron flux, because it is ordinarily not possible to count all recoil protons. However, the efficiency dose provides a convenient starting point for further calculation.

$$N = N_0 \frac{n_H \sigma_H}{n_H \sigma_H + n_c \sigma_c} \left[1 - e^{-(n_H \sigma_H + n_c \sigma_c)L} \right] \quad \cdots\cdots\cdots\cdots (3)$$

where N is the number of recoil protons produced in the scintillator ; No, the neutron flux (n/cm²) ; n_H, the number of hydrogen atoms per cm³ of the scinillator ; n_c ,the number of carbon atoms per cm³ of the scintillator ; σ_H, the neutron-proton scattering cross sect-

ion for 14.1 MeV neutron ($=0.695$ barns); σ_c, the neutron-carbon scattering cross section for 14.1 MeV neutron ($=1.35$ barns) and L, the length of the scintillator ($=2.59$ cm).

National scintillite was used as the scintillator and its chemical composition was C: 92% and H: 8%, and the density was 1.05 g/cm^3.

The efficiency of the scintillite for a single scattering was calculated to be 0.0795/cm^2. The calculation was carried out under the assumption that only single scattering occurs and scintillite consists of carbon and hydrogen atoms.

One obious limitation in this method is that only single scattering events were considered. Recoil protons can, of course, be produced by neutrons which have been scattered one or more times by carbon and hydrogen. The corrections for these effects are difficult to calculate accurately. The contribution of second scattering events is estimated as follows:

$$E_2 = E_1 \ (1 + N_2/N_1) \quad \cdots\cdots\cdots\cdots(4)$$

where E_2 is the efficiency of scintillator, including double scattering; E_1, the efficiency of scintillator for single scattering; N_2, the number of recoil protons produced in double scattering process; N_1, the number of recoil protons produced in a single scattering process.

However, it is found that in many cases the efficiency calculated from equation (4) is almost equal to the efficiency calculated under the assumption that carbon does not collide with neutrons at all. Efficiency is simply

$$E_0 = 1 - e^{-n_H \sigma_H L} \quad \cdots\cdots\cdots\cdots(5)$$

thus the efficiency of the scintillator used, including double scattering, was calculated to be 0.0859/cm^2. This value is about 8 percent higher than that for single scattering.

The loss of protons from the end and side of the scintillator bring rise to increase at low energies and to decrease at high energies but the total number of recorded protons is not affected. This effect in the scintillator used in this study is so small that it can be ignored.

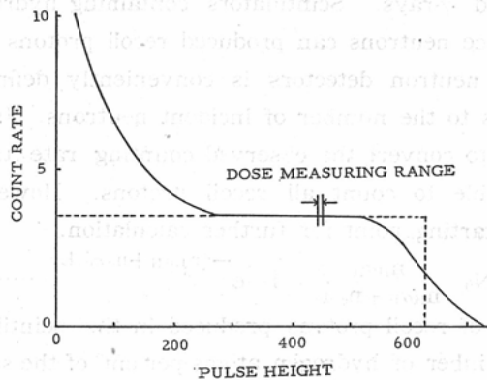Figure 6 shows the pulse height spectrum of recoil protons. The proton energy

Figure 6. Pulse height distribution curve of recoil protons

spectrum due to single scattering must be rectangular, but the increase of count rate in the lower energy level was demonstrated by the effects of recoil carbon nucleus and γ-ray from activated substance or inelastic collision, etc. In order to avoid the error arising from these effects, the neutron flux must be calculated from counts within the dotted line in Fig. 6. In our laboratory, total counts within the dotted line have been estimated from counts within the range of $450+6.25$. In this sense, it is desirable to use a scintillation counter equipped with a pulse height analyser.

III Comparison between activation method and scintillation method

The merits of the activation method are: 1) There is no effect from γ-ray and recoil carbon nucleus. 2) The sample can be fixed in a similar position and made in a similar volume to irradiated animals. The disadvantageous points of this method are: 1) The irradiation dose and the dose rate during irradiation are unknown and the total irradiation dose cannot be controlled, although they may be roughly estimated from the current of deuteron beam. 2) The value to be used as the nuclear reaction cross section of ^{32}S (n,p)^{32}P, that is 0.30 or 0.37 barns, will largely affect the accuracy of the dosimetry.

The scintillation method has the following advantages: 1) The irradiation dose and the dose rate during irradiation can be calculated from the counts registered by the scintillation counter and a given dose can be irradiated to animals. 2) The neutron-proton scattering cross section has been determined with considerable accuracy. The demerits of this method are: 1) Because of various limitations the scintillator cannot be fixed in a similar position with animals. Therefore, the scintillator was placed 100 cm from the target and the neutron flux was calculated with the assumption that it attenuates by the inverse square of distance. 2) The scintillator is sensitive to γ-ray and to recoil carbon nucleous. These effects may be minimized by the use of a pulse height analyser.

Table III shows radiation doses received by mice at 5 cm from the target. The

Table III Comparison between activation method and scintillation method

Exptl No.	Scintillation dose (rad)		Activation dose (rad)	
	(I) Single collison $\varepsilon=0.0795$	(II) Multiple collision $\varepsilon=0.0859$	(III) $\sigma=0.30$	(IV) $\sigma=0.37$
1	392	347	405	329
2	352	311	362	294
3	316	278	311	256
4	235	208	240	195
5	195	164	193	156
6	557	492	660	535
7	465	411	572	464
8	400	370	490	397
9	510	472	537	436
10	500	463	570	462

Data shows irradiation dose of mice at 5 cm from target, scintilla-tor-target distance : 100 cm, sulfur detector-target distance : 5.0 cm~11.0 cm

doses were measured by the scintillation method and the activation method. First column
(I) in Table III shows the values calculated under the assumption that only single
scattering occurs. Second column (II) shows that of double scattering. Third (III) and
fourth column (IV) show values of the activation method calculated using 0.30 and 0.37
barns as the nuclear reaction cross section, respectively.

For the conversion factor of rad per neutron in animals, 6.7×10^{-9} calculated according
to Randolph[9] under the assumption that only single scattering occurs was used. The
doses in Table III were obtained by flux multipled by 6.7×10^{-9}. The true conversion
factor of rad per neutron appears to be slightly higher than 6.7×10^{-9}, because in animals
multiple scattering events also occur. Therefore, the doses of II, III and IV in Table
III are slightly lower than the true values.

In the single scattering scintillation method for the neutron flux a slightly higher
value was estimated than the true flux.

When the elemental composition and volume of the scintillator is similar to that of
irradiated animals, it is possible that the error of the flux estimation and the error of
the conversion factor of rad per neutron will mutually compensate with each other.

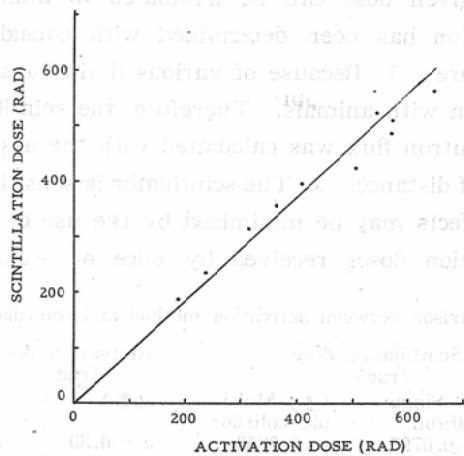Fig. 7 shows the curve which relates the activation dose (III) to the scintillation dose

Figure 7 Relationship between activation dose and scintillation dose
scintillation: single collision
activation: $\sigma_s = 0.30$ barns

(I). The dose ratios of I/III, I/IV, II/III, II/IV, I/II and III/IV were 0.925, 1.140, 0.856,
1.056, 1.081 and 1.233, respectively. The highest dose was obtained in the activation
method (III) and the lowest dose was obtained in the activation method (IV).

The RBE of neutron to X-ray for mortality in mice (ddN uniform strain) was found
to be 1.14 by Sawada of our laboratory. The dosimetric method used in this study was
the scintillation method (I). The estimated RBE changes; that is 1.23 with the use of
the scintillation method (II), 1.05 in the activation method (III) and 1.30 in the activation
method (IV).

Summary

1) The main activation product in irradiated sulfur was identified to be radioactive phosphorous ^{32}P.

2) The competing reaction was observed. Although the competing product could not be identified, it is considered to be radioactive silicium ^{31}Si.

3) The difference of the activity in each sample irradiated under the same condition was less than $\pm 10\%$, but in the absolute neutron flux measurement, the nuclear reaction cross section of ^{32}S (n,p) ^{32}P reaction strongly affects the accuracy of the dosimetry.

4) The activity of the samples at given distances from 5 cm to 11 cm from the target attenuates by the inverse square of distance.

5) The counting efficiency of the scintillator for neutrons was $0.0795/cm^2$ for single scattering and $0.0859/cm^2$ for double scattering.

6) The ratio of scintillation dose to activation dose was 0.925 (single scattering) and 0.856 (double scattering) when 0.30 barns was used as nuclear reaction cross section of ^{32}S(n,p) reaction and 1.140 (single scattering) and 1.056 (double scattering) when 0.37 barns was used.

7) The error of dose estimation due to the dosimetric method was within $\pm 25\%$.

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